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Growth and Property Development of Convection Pass Deposits in Recovery Boilers

Final Project Report

Larry L. Baxter, Richard A. Wessel, W. James Frederick Jr., and Steve J. Lien

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GROWTH AND PROPERTY DEVELOPMENT OF CONVECTION PASS DEPOSITS IN RECOVERY BOILERS

Final Project Report

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Abstract

As part of the U.S. Department of Energy (DOE) Office of Industrial Technologies (OIT) Industries of the Future (IOF) Forest Products research program, the mechanisms of particle deposition and properties of deposits that form in the convection passes of recovery boilers were investigated. Research from experimental facilities at Sandia National Laboratories, the Institute of Paper Science and Technology (IPST), and the University of Toronto (UofT) was coordinated into a single effort to define the controlling mechanisms and rates of deposition. Deposition rates were recorded on a volumetric and mass basis in a Sandia facility for particle sizes in the range of 0.1 to 150 μm. Deposit thickness, mass, spectral emissivity, thermal conductivity, surface temperature, and apparent density were monitored simultaneously and *in situ* on instrumented probes that allow determination of heat flux and probe surface temperature. Particle composition and mass deposition rates were also recorded in a UofT facility for particle sizes in the range of 100 to 600 μm. These measurements allowed determination of the liquid content and sticking efficiency of carryover particles that inertially impact on a deposition probe. In addition, information on particulates, stable gas species, gas temperature and velocity were obtained from field tests in an operating recovery boiler. The results were used to develop algorithms appropriate for use in computer codes that simulate recovery boilers. Representative calculations were performed using B&W's comprehensive recovery boiler model to demonstrate the use of the algorithms in such computer codes. Comparisons between observations in commercial systems and model predictions were made to identify algorithm strengths and weaknesses.

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Acknowledgments

This project succeeded because of the contributions of a large number of individuals at Sandia, the Institute of Paper Science & Technology, McDermott Technology, and the University of Toronto, as well as among our industrial hosts and collaborators at Weyerhaeuser, International Paper, and Babcock & Wilcox. The assistance of the American Forest and Paper Association (AF&PA) and the Department of Energy was also important.

Don Hardesty, Sandia manager, assisted with the original project proposal and project management during the course of the project.

Bob Roscoe and Bruce Stone of Weyerhaeuser assisted with various aspects of project planning, mill coordination, boiler testing, and data interpretation at Longview recovery boiler.

Tertalissa Lind, Mark Rumminger, Gian Sclippa, John Nelson, and Tim Buteau, assisted with measurements at Weyerhaeuser's Longview boiler. We thank Pirita Mikkanen and Esko Kauppinen for many fruitful discussions and advice on aerosol sampling, and Anna Moisala for help in SEM analysis. Funding provided by VTT Chemical Technology is gratefully acknowledged.

Scott Sinquefield of Institute of Paper Science & Technology carried out previous experiments on Sandia's Multifuel Combustor (MFC), which provided guidance for the current investigation, and contributed to the analysis of fume deposition rates. Terttalissa Lind and Allen Robinson operated the MFC and conducted measurements of deposit thermal properties.

Honghi Tran and the faculty of the Pulp & Paper Centre, University of Toronto, provided cost-share support of this project, through collaborative research on fume sintering and carryover deposition. Reyhaneh Shenassa conducted carryover deposition measurements with the Entrained Flow Reactor (EFR). Nanan Pathania, Nader Mahinpey, Ashgar Khalaj, and Professor David Kuhn are acknowledged for the experimental and numerical investigation of carryover composition in the EFR. Saied Kochesfahani conducted independent field measurements of ISP in recovery boilers.

The efforts of all of these individuals, and any we have failed to mention here, are appreciated and were essential to the successful completion of this project.

Executive Summary

This report documents the results of work performed on a project entitled "Growth and Property Development of Convection Pass Deposits in Recovery Boilers," which was funded by the U.S. Department of Energy (DOE), with industrial cost-share, through the auspices of the Forest Products focus area within the Office of Industrial Technologies (OIT) Industries of the Future (IOF) program. Technical evaluation of the proposed work and of the progress during the course of the project occurred through the American Forest and Paper Association (AF&PA) Agenda 2020 Energy Performance Committee.

The purpose of the project was to investigate the mechanisms of particle deposition and properties of deposits that form in the convection passes of recovery boilers. The work was a collaborative and multidisciplinary effort involving Sandia National Laboratories, McDermott Technology, Inc. (MTI) affiliated with Babcock &Wilcox (B&W), the Institute of Paper Science & Technology (IPST), and the University of Toronto (UofT). The project was divided into four tasks: (1) Particle Formation and Properties, (2) Deposit Characterization, (3) Algorithm and Model Development, and (4) Model Validation.

The objective of the first task was to characterize the rates and mechanisms of particle generation and to determine the properties of entrained particles under commercial conditions. This was initially accomplished by extending experimental work on IPST's char bed reactor, to quantify the formation rate of intermediate sized particles during char bed burning, and to evaluate the impact of char preparation techniques on experimental results. These experiments confirmed earlier results and demonstrated that intermediate-sized particles were not an artifact of the laboratory char formation process. More definitive information on particle size distribution and concentration in the flue gases was obtained from extractive particle sampling in the upper furnace and convection pass of Weyerhaeuser's Longview WA recovery boiler. Measurements of stable gas composition, gas velocity and temperature were also obtained in essentially every accessible port in the boiler. Fume particle size distributions peaked at about 0.7 μm physical diameter, and fume particle concentration ranged from 8.9 to 11.7 g/Nm^3; neither the size nor the concentration of fume particles changed significantly through the convection pass. Intermediate-sized particles (1-100 μm) could not be reliably sampled, although crude estimates of their concentration were made.

The objective of the second task was to characterize deposit growth mechanisms and rates and to measure the development of critical deposit properties under commercial-scale conditions. Time-resolved deposition rates were recorded on a volumetric and mass basis in a Sandia facility for particle sizes in the range of 0.1 to 150 μm. Deposit thickness, deposit mass, spectral emissivity, effective thermal conductivity, surface temperature, and apparent density were monitored simultaneously and *in situ* on instrumented probes that allow determination of heat flux and probe surface temperature. Experiments on fume deposition indicate that deposit mass accumulation increases linearly with time and is independent of the temperature gradient – results that are inconsistent with the classical explanation of thermophoresis as the deposition driving force. Particle composition and mass deposition rates were also recorded *in situ* in a

University of Toronto facility for particle sizes in the range of 100 to 600 μm. University of Toronto measurements allowed determination of the liquid content and sticking efficiency of carryover particles that inertially impact on a deposition probe.

As part of the second task, the sintering and densification of fume deposits were investigated at IPST and University of Toronto at conditions that are representative of a recovery boiler. One objective of the sintering experiments was to evaluate the impact of gas composition on the sintering rates of electrostatic precipitator (EP) dusts. Gas composition was found to have little or no effect, except in the unlikely event that SO_2 concentrations exceeded 1.0%. Another objective of sintering measurements was to evaluate the impact of deposit structure on sintering rates for low density deposits. The analysis of scanning electron microscope (SEM) photomicrographs and chemical composition of deposits, formed *in situ* in an operating recovery boiler, provided detailed information about the structure, particle size, porosity, sintering behavior, and composition of deposits in the superheater, boiler banks, and economizer.

The objective of the third task was to develop algorithms that relate the data collected in the first two tasks to recovery boiler operating parameters and other observables. Sub-models describing particle formation, particle deposition, sintering and deposit properties were developed during the course of this investigation. Of the three classes of particulate (carryover, ISP, and fume), carryover formation is perhaps the best understood and most accurately modeled. Fume formation is not as well understood due to the complexity of vaporization-condensation, aerosol dynamics, and sulfur scavenging from the flue gas. ISP formation is the least understood of the three classes of particulate, and rational sub-models for ISP formation have not been developed, with the exception of a model that describes physical ejection of ISP during pyrolysis of black liquor drops. Available sub-models for particle deposition are applicable over the entire range of particle sizes 0.1 to 1000 μm; however, there are still large uncertainties in the ability to predict fume deposition rates based on sub-models for thermophoretic deposition alone. Both commercial operation and detailed laboratory experiments suggest that fume deposit mass increases proportionately with time, and that other mechanisms such as surface renewal may be involved. A correlation for fume deposition rate was developed for a single-tube in crossflow; however, the correlation has not been generalized for convection pass tubes of a recovery boiler. Rational sub-models for deposit thermal properties (emissivity and thermal conductivity, as a function of particle size, deposit structure and density) have been formulated, but depend upon sub-models for fume deposition rate.

The objective of the fourth task was to incorporate the algorithms derived in the third task into a comprehensive computer code, and demonstrate an improved ability to predict ash behavior in recovery boilers as a function of fuel composition, boiler design, and operating conditions. Available sub-models were implemented into B&W's comprehensive recovery boiler model, COMO-PR. Comprehensive modeling of black liquor combustion and ash deposition was performed for three recovery boilers, two of which were test sites for particle sampling and furnace probing. Comprehensive modeling of particle formation and deposition was useful as a forensic tool for explaining experimental observations and evaluating particle formation and deposition sub-models. However, modeling has not yet been developed to the level of sophistication needed to predict the behavior of convection pass deposits accurately. Although ash deposition predictions are insightful, they cannot be analyzed or interpreted with confidence until rational sub-models for ISP formation and destruction are developed. Sub-models are also needed to describe experimentally observed fume deposition rates, deposit properties, and potential interaction with ISP particles. Field data gathered on this project (and a related project focused on ISP) will be valuable for testing new algorithms, for validating modeling results, and for improving our understanding of particle formation and deposition processes.

Background

This report documents the results of work performed on a project entitled "Growth and Property Development of Convection Pass Deposits," which was funded by the U.S. Department of Energy (DOE), with industrial cost-share, through the auspices of the Forest Products focus area within the Office of Industrial Technology (OIT) Industries of the Future (IOF) program. This project was recommended for funding in 1997 through the Energy Performance Subcommittee of the American Forest and Paper Association (AF&PA) Agenda 2020 program and, subsequent to project funding in late 1997, technical progress was monitored through regular meetings of this same subcommittee.

This project was motivated by the ubiquitous problem of excessive deposit formation in the convection pass of U.S. (and indeed, all North American) kraft pulp mill recovery boilers. These deposits reduce the energy efficiency of these boilers, limit the pulping capacity of many U.S. kraft pulp mills, and often result in boiler pluggage and lost capacity. The fuel that is burned in the Tomlinson furnace is known as strong black liquor, and typically consists of 20 wt-% sodium (on a dry basis) in a mixture of lignin-derived organics, water, and sulfur compounds [Hupa 1997]. The primary functions of recovery boilers in pulp and paper mills are: 1) to recover inorganic chemicals from black liquor for recycling through the mill; and 2) to transform chemical energy in the organic portion of black liquor to thermal energy for use in steam or electrical power generation. In the combustion process, inorganic material is released from the fuel, incorporated into the combustion gases as both particles and vapors, and deposited on boiler surfaces. Heat transfer distribution in furnaces depends in large measure on the overall quantity and thermal transport properties of the inorganic material present. Deposit formation in the convection pass represents one of the major operation and design limitations for the boiler.

The traditional view of particle generation in recovery boilers classifies particles according to two important classes of particles whose existence is readily apparent in the furnace flow: carryover particles and fume [Tran 1997]. Carryover particles, which are generally considered to be 100 microns or greater in diameter, originate from black liquor droplets that burn out in flight or from entrainment of particles from the char bed at the bottom of the recovery boiler. Their mechanisms of formation and influence on deposition are relatively well understood and continue to be investigated. Fume particles, which are less than 1 micron in diameter, are formed by condensation of inorganic salts that are vaporized during black liquor combustion. Their mechanisms of formation are conceptually understood and their rates of deposition seem to be well characterized, if not completely understood.

In recent years, several studies [Verrill et al. 1994; Sinquefield 1998; Sinquefield et al. 1998b; Mikkanen et al. 1999; Baxter et al. 2000; Kochesfahani and Tran 2000; Kochesfahani et al. 2000; Lind et al. 2000; Baxter et al. 2001b; Lind et al. 2001; Mikkanen et al. 2001; Holve et al. 2001; Shaddix and Holve 2004; Ip et al. 2004] have highlighted the potential importance of particles with sizes 1 to 100 microns, between the two well-studied classes, carryover and fume. For lack of a better name, or understanding of their origin in boiler, these particles have come to be known as intermediate-sized particles or ISP. The relative contribution of this class of particles to deposit formation in different regions of the boiler is highly dependent upon the particle size distribution, chemical composition, and overall concentration in the flue gas. In order to understand and predict the effect of ISP on deposit formation, the characteristics of such particles, in commercial furnaces, needs to be measured, and the chemical and physical mechanisms that result in ISP formation need to be identified.

Particle formation and deposition in recovery boilers was the focus of two projects that were recently completed for the U.S. Department of Energy, Industrial Technologies Program in Forest Products. This report summarizes the results of the first project, “Growth and Property Development of Convection Pass Deposits in Recovery Boilers”. The goal was to conduct a systematic and fundamental investigation of structural, optical, and transport properties of entrained particles and ash deposits representative of those found in commercial scale recovery boiler convection passes. A separate report [Shaddix et al. 2004b] describes the results of the second project, “Intermediate-Sized, Entrained Particles: Characterization, Formation and Control,” which is closely related to and an outgrowth of the first project. The goal was to quantify the concentration of ISP in recovery boilers, identify the mechanisms and rates of ISP formation and deposition, and propose methods to control their effects on boiler operation. A summary of both projects was presented at the International Chemical Recovery Conference [Wessel et al. 2004]. Detailed results of both projects are mainly documented in the form of technical publications that are referenced throughout the final reports and the project summary conference paper.

Particle Generation and Properties

The initial focus of investigation was on deposit formation in the back sections of the convection pass (or backpass) in a recovery boiler, which was believed to be caused primarily by fume particles. However, as described in the previous section, there were several independent indications that ISP is also generated in a recovery boiler and entrained by the flue gas into the backpass. Specifically, the fume deposition experiments conducted as part of this project produced deposits more characteristic of those found in recovery boilers (dominant growth on leading edge, etc.) when super-micron-sized particles were included in the flow [Sinquefield 1998; Sinquefield et al. 1998b; Baxter et al. 2001a; Baxter et al. 2004a; Baxter et al. 2004b]. Therefore, the generation and properties of both fume and ISP were considered.

Laboratory Measurements

Three sources of ISP were considered in this work: the char bed, direct formation from a splatter plate, and ejection during drying and devolatilization. Much of this work was continued in a closely related project involving most of the same investigators but focused on ISP specifically. This is summarized separately [Shaddix et al. 2004a; Shaddix et al. 2004b; Wessel et al. 2004], but a few of the highlights and early indications of ISPs are described here.

One investigation of ISP (1-100 μm) from a char bed inspired by Sinquefield’s observations used simulations of char bed burning using IPST’s char bed reactor [Kochesfahani et al. 2000]. These experiments demonstrated that ISP represent as much as 8% of the initial char mass, and account for 40 to 80% of the total particulate mass collected. However, one issue remained unresolved with these experiments: whether these particles were due to the mechanical process that was used to prepare the char. Therefore, experimental work on char bed burning was continued using a new method of generating the char. The char was produced by pyrolizing black liquor at 750ºC under a nitrogen atmosphere, inside the IPST char bed reactor, prior to performing the combustion test. These experiments confirmed earlier results [Kochesfahani et al. 2000] and demonstrated that intermediate sized particles were not an artifact of the char formation process. It was not clear, however, how much ISP was formed, with estimates ranging from essentially 0 to nearly 15% of liquor dry solids depending on how the data were interpreted. The data from this work, as well as additional char bed burning experiments with a new reactor design, are reported elsewhere [Lien and Verrill 2004].

No definitive indication of ISP formation from spray nozzles has been observed. Several experiments have been attempted, but in most cases the optical resolution was insufficient to detect ISP-sized particles. Inferences from the data that were obtained suggest that the viscosity and drying of liquor injection may argue against ISP formation, but this remains an open issue.

ISP formation during droplet reactions has been quantified. Early indications [Verrill et al. 1994] of ejection of inorganic particles are corroborated, in part, by similar observations for other fuels [Baxter 1990; Baxter and Mitchell 1992]. Subsequent work funded in part by a closely related project confirmed the early indications that small amounts of ISP are formed during drying and devolatilization, but also suggested that the majority of ISP is formed during high-temperature char oxidation. The amount of ISP from this process ranged from < 1% to 2% of dry solids content and scaled with the time the char particle remains at high temperatures (which scales with solids loading, for particles of the same initial size) [Baxter et al. 2004b; Ip et al. 2004; Shaddix et al. 2004a].

Field Measurements

Field measurements conducted under this project confirmed the presence of ISP in recovery boilers [Lind et al. 2000; Baxter et al. 2001b]. These measurements focused on fume deposition but found several indications of ISP in addition to fume in the upper section and convection pass of a furnace. Analyses included extracted particles through water-cooled probes, gas compositions, temperatures, impaction analyses on probes, and gas velocities. Independent field measurements conducted by the University of Toronto confirmed the presence of intermediate-sized particles in operating recovery boilers based on impaction measurements on probes [Kochesfahani and Tran 2000]. This supported the earlier conclusion that suggested that deposit structures, of the type observed in recovery boilers, could not be generated from fume alone [Sinquefield et al. 1998a]. It was also consistent with results from IPST that suggested that intermediate-sized particles are generated during char bed combustion. These factors were the motivation for sampling the particulate in an operating recovery boiler in the present work, and for proposing a separate project focused on intermediate-sized particles.

Field tests were conducted at Weyerhaeuser's Longview (WA) recovery boiler to establish the existence of intermediate-sized particles (1-100 μm) in an operating recovery boiler, and to determine fume and carry-over particle size and concentration. Particle samples were collected from two locations in the furnace and in the generating bank. Size distributions and concentrations of fume particles were determined with a Micro-Orifice Uniform Deposit Impactor (MOUDI) using a water-cooled, N_2-quenched probe. Particle samples were also collected inside the furnace for morphology characterization. In addition to particle sampling, measurements of gas composition, velocity, and temperature were obtained from essentially every accessible port in the boiler [Lind et al. 2000; Baxter et al. 2001b].

Three kinds of intermediate-sized particles were observed with SEM: droplets of 50-1000 μm in diameter (after impaction on a foil), agglomerates, and roughly spherical particles 2-10 μm in diameter. Droplets were only found in the furnace, but the other two particle types were found at all locations. The main compounds in the particles were Na and SO_4.

The concentration of the fume particles was 8.9-11.7 g/Nm^3, and the mean aerodynamic diameter 1.1 μm. The size and concentration of the fume particles changed only slightly from one measurement location to another and from one day to another, indicating that the fume particles formed in the furnace before the first sampling port and only a small fraction of the particles collected on convection pass surfaces. The data also indicate that fume size and concentration are relatively insensitive to changes in the process conditions. As expected, the main compounds in the fume particles were Na and SO_4, with smaller amounts of K and Cl.

The methods and results of particle sampling during the December 1999 tests at Longview recovery boiler were documented in several technical publications [Lind et al. 2000; Baxter et al. 2001b; Lind et al. 2001]. A separate project that focused on intermediate-sized particles, including subsequent measurements using *in situ* laser diagnostics at the Longview recovery boiler and one other recovery boiler, is described by Shaddix et al. [2004b].

Deposit Characterization

The Multifuel Combustor (MFC) at Sandia National Laboratories Combustion Research Facility (Figure 1) was developed to measure deposition rates and deposit properties *in situ* and as a function of time for a variety of fuels. The reactor is a down-fired, small pilot-scale, turbulent flow combustor that allows for gas and particle temperature histories to be varied over the range of conditions commonly experienced in commercial combustors. A temperature controlled probe was placed at the exit of the furnace flow and deposits were accumulated on it. The probe typically rotated on its axis to grow a physically symmetric deposit. As the deposit grows, optical techniques were used to measure its spectral emittance, surface temperature, and thickness while thermocouples imbedded at the surface of the rotating probe and in the metered stream of internal cooling air were used to monitor the deposit interior surface temperature and the total heat flux, respectively.

Experimental measurements on black liquor ash deposits were initially conducted in the MFC as a result of a DOE-sponsored collaboration [Sinquefield 1998; Sinquefield et al. 1998a]. Deposition of fume and ISP on tube surfaces was characterized under laboratory conditions that

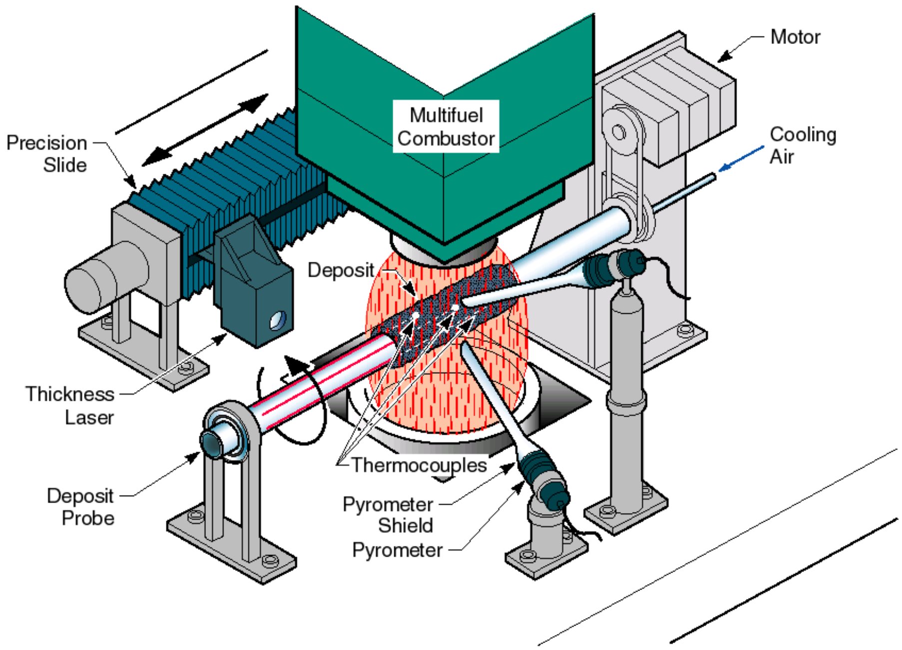


Figure 1. Sandia's Multifuel Combustor and deposition probe for characterizing deposit thermal conductivity and emissivity.

closely simulated the range of conditions used in commercial recovery boilers. The objective of the measurements was to determine the mechanisms and rates of deposition, while independently varying the particle size distribution, particle loading, tube Reynolds number, tube Stokes number and gas temperature. The primary conclusions of this work were that the fume depositionmechanism does not appear to be controlled by thermophoresis; fume and ISP deposit mass increases linearly with time under a wide variety of conditions, and deposition rates remain largely unaffected by rising deposit surface temperature with increased amount of deposit. However, deposition rates were affected (slightly) by initial surface temperature. Alternative theories and calibrated predictive techniques for describing fume deposition have recently been proposed [Baxter et al. 2004a] that scale proportionally to time and that account for over 95% of the variation in the data.

The thermal conductivity and emissivity of black liquor ash deposits were also measured in the MFC as part of this project. Detailed results of this work and the techniques used to determine the properties were published separately [Bernath et al. 1998; Baxter et al. 2001a; Robinson et al. 2001a; Robinson et al. 2001b; Baxter et al. 2004a; Baxter et al. 2004b], and the key results are summarized here. The experimental approach and facilities preserved the dominant aspects of deposit structure while allowing for quantitative, *in situ* determination of these properties. The radiation spectra from deposits, useful for determination of surface chemical composition, are dominated by peaks associated with carbonates and sulfates. Average (Plank-weighted) total emissivities for fume and larger particles were calculated from this data as shown in Figure 2. Experimental results show that total emissivity values tend to decrease with temperature, and increase with particle size. The temperature dependence is not an indication of sintering or other physical or chemical changes. It is predominantly caused by shifts in the most active spectral regions of black body radiation with temperature. Changes with size are consistent with emissivities from particulate surfaces, with smaller particles offering more surface area from which light scatters and hence producing more reflective deposits.

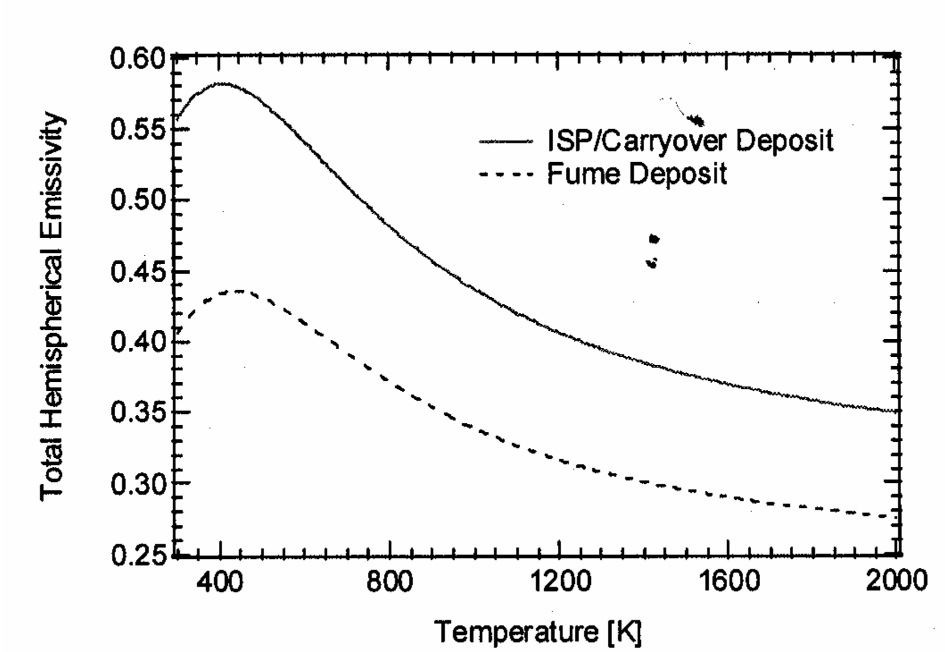
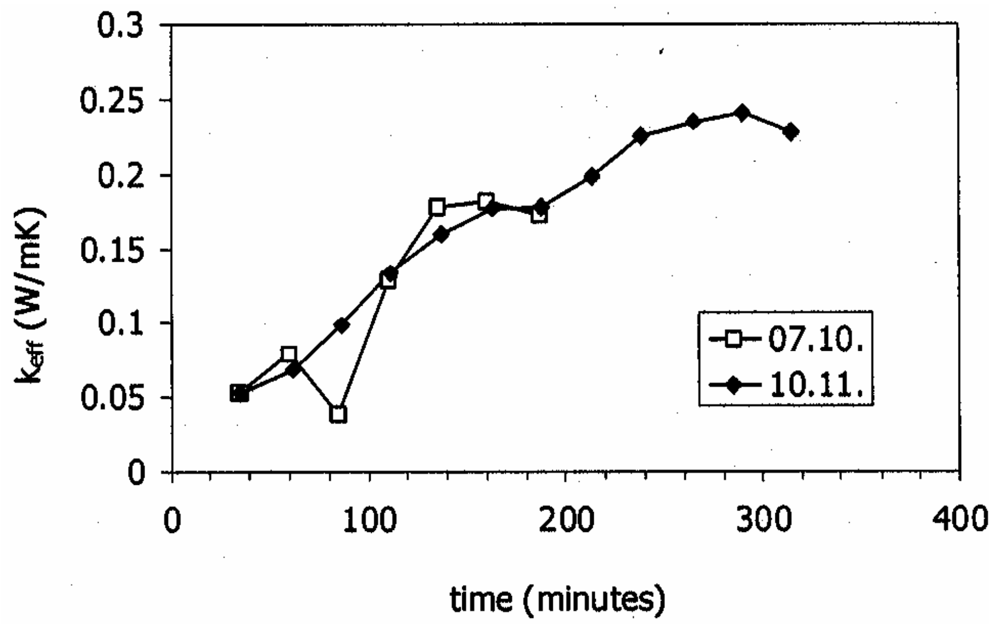


Figure 2. Total hemispherical emissivity for fume and larger particles as a function of absolute temperature.

The thermal conductivity of fume deposits was shown to increase as sintering and deposit temperature increased with time (Figure 3). These experiments demonstrated a dependence of deposit strength and thermal conductivity on the structure and porosity of the deposit. A theoretical model was developed to describe thermal conductivity in terms of a structural parameter and the upper and lower limits for solid phase and gaseous phase heat conduction. Deposit strength and thermal conductivity were found to correlate well with the same structural parameter [Robinson et al. 2001a; Robinson et al. 2001b].

Carryover Deposition

An Entrained Flow Reactor (EFR) at the University of Toronto (Figure 4) was designed to measure carryover particle composition and deposition. The experimental apparatus consists of a particle feeder, a gas combustion unit, a long vertical heating section, a sampling and optical diagnostic section, a gas exhaust system, and a data acquisition and control system. The particle feeder consists of a 0.15 m by 1.2 m belt conveyor that transports particles at a mass flow rate of 0.5 to15 g/min to the top of a water-cooled particle injector which introduces the particles into the first furnace of the reactor. The heating section is an assembly of five split-shell tube furnaces, which form a continuous vertical unit. Each furnace can be heated electrically to a maximum temperature of 1350ºC, and the temperature of each furnace can be controlled



independently. A 0.18 m ID mullite tube is placed at the center of each furnace and there is a 0.1

Figure 3. Typical development of thermal conductivity with increased sintering temperature / time for black liquor deposits.

m non-heated insulated space which accommodates an observation /sampling port between adjacent furnaces. The gas combustion unit is equipped with a gas burner that has a maximum heat input of 44 kW. Natural gas is burned at a flow rate of up to 4.5 SCMH, and the combustion gas is tangentially mixed with dilution air to produce the desired gas volume and temperature (up to 1200 ºC). The gas is exhausted from the bottom of the reactor by a centrifugal induced draft fan.

The EFR was initially used to simulate conditions experienced in the convection section of a recovery boiler, and to study synthetic carryover deposition as a function of liquid content [Shenassa et al. 1999; Shenassa 2000; Shenassa et al. 2003]. Particle sizes in the range from 100 to 600 μm were investigated. The experimental study, and supporting numerical results, demonstrated that liquid content is the most important parameter determining sticky temperature and deposition rate of carryover particles. The liquid content and sticky temperature can be calculated based on multiphase chemical equilibrium predictions for alkali salt mixtures containing sodium, potassium, chloride, carbonate and sulfate [Tran et al. 2002]. Sticky temperature generally decreases with increasing chlorine and potassium content. However, the composition of the carryover must be known *a priori* to predict the sticky temperature.

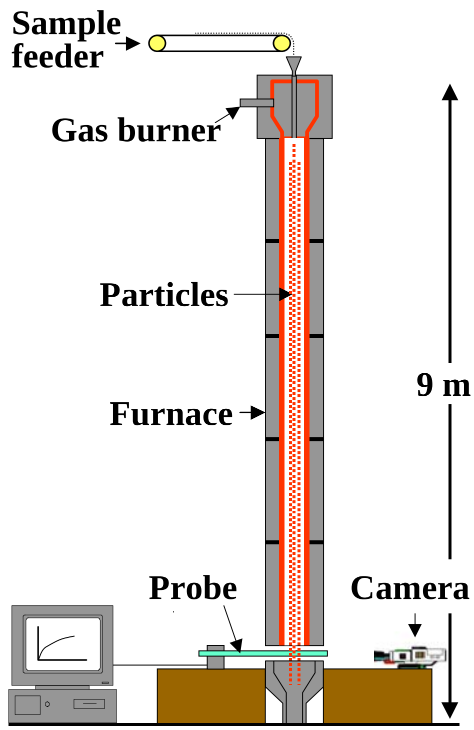


Figure 4. Schematic of Entrained flow Reactor at University of Toronto.

As an extension of this work, the EFR was used to study the composition of carryover particles obtained from burning dried black liquor [Pathania 2002; Khalaj et al. 2004]. The key result of these studies was significant depletion of chlorine and potassium in carryover particles, via NaCl and KCl vaporization, which decreased with particle size and increased with gas oxygen concentration. The depletion of chloride was roughly 3 times greater than the depletion of potassium, due to the fact that chloride vaporizes as both NaCl and KCl, while potassium vaporizes only as KCl. Numerical modeling demonstrated similar trends in carryover composition for the EFR and a recovery boiler [Wessel et al. 2002]. Therefore, higher oxygen concentrations and more intense burning of carryover particles leads to greater depletion of chlorine and potassium, higher sticky temperature, and reduced deposition of carryover.

Sintering and Densification of Fume Deposits

Submicron fume particles are entrained in the flue gas, at typical concentrations of 10 to 20 g/Nm^3 [Mikkanen et al. 1999; Lind et al. 2000; Baxter et al. 2001b], enriched in chlorine and potassium, and deposited on tubes throughout the convection pass of recovery boilers. The low temperature melting behavior of the dust particles cause the deposits to sinter and harden, leading to tenacious deposits that are sometimes difficult to remove by sootblowing. Deposit formation in the convection pass reduces the thermal efficiency of the boiler, increases flue gas draft loss and blockage, and is a persistent and costly problem for boiler operation.

Sintering and densification of fume deposits has been extensively studied at the Institute of Paper Science & Technology and the University of Toronto for electrostatic precipitator (EP) dusts with varying composition, from a number of boilers [Techakijkajorn et al. 1999; Frederick et al. 2004, Duhamel et al. 2002]. These sintering studies, on high- and low-density pellets made from compressed EP dust, were performed under a nitrogen atmosphere. One of the objectives of the present research at IPST was to understand sintering rates in a gas environment that is more representative of that found in the convection pass of a recovery boiler. Therefore, tests were performed to examine the impact of gas composition (O_2, H_2O, CO_2, and SO_2) on the sintering rate of EP dusts [Lien et al. 2004]. Sulfur dioxide was found to have a large impact on sintering at SO_2 concentrations of 1.0%, but little or no effect for SO_2 concentrations of 0.1% or less. None of the other gases had a statistically significant effect.

Another objective was to investigate the impact of deposit structure on sintering rates, for low-density deposits that are more representative of a recovery boiler. The analysis of scanning electron microscope (SEM) photomicrographs and chemical composition of deposits formed *in situ* in an operating recovery boiler provided detailed information about the structure, particle size, porosity, sintering behavior, and composition of the deposits in the superheater, boiler banks, and economizer [Frederick and Vakkilainen 2003]. A theoretical model for sintering of recovery boiler deposits was formulated, which incorporates different size-scales for sintering of poly-disperse particles and branched dendritic structures in low apparent density deposits. In theory, both the rate and extent of sintering depend upon the macro-scale structure. Rate equations were developed that describe initial stage (neck growth) sintering based on a rigorous geometric model of Lunden [1995]. These equations described the rate of mass transport to the neck region and the geometry of spheres and neck region for a two-sphere system for pure NaCl, but lacked available date for diffusion coefficients and surface energy for particles containing alkali chloride, carbonate and sulfate.

A simpler, yet more empirical form of sintering algorithms was developed that is applicable for a greater range of conditions and is more tractable for engineering calculations [Frederick et al., in press]. These algorithms rely on a correlation between the sintering rate constant for evaporation-condensation of NaCl and KCl and the first melting temperature of the dust. The correlation is valid for the initial stage of sintering, during which the greatest shrinkage and densification of the deposit occurs.

Deposition and Thermal Property Algorithms

New sub-models for describing particle formation, particle deposition, and deposit properties were developed during the course of this investigation. Many of these are summarized in other documents [Sinquefield et al. 1998a; Baxter et al. 2000; Baxter et al. 2001a; Lind et al. 2001; Baxter et al. 2004a]. Table 1 summarizes the status of sub-model development based on theoretical understanding, embodiment of theory and experiments in the form of mathematical algorithms and correlations, and validation with experimental data. Sub-models with a maturity level of 2 or above are considered suitable for implementation in a comprehensive CFD code. Sub-models with a maturity of 1 or less either do not exist or are considered as having insufficient experimental data and/or foundation in physics and chemistry to describe the behavior in a recovery boiler.

Of the particle formation processes, carryover formation is perhaps best understood and most accurately modeled. However, additional work is needed to characterize the inorganic reactions needed to predict carryover composition, liquid content, and sticky temperature. Fume formation is not as well understood due to the complexity of vaporization-condensation, aerosol dynamics, and sulfur scavenging from the flue gas. Sub-models for fume formation are perhaps capable of predicting the quantity of fume, but have greater difficulty predicting fume composition and chemical reactions.

ISP formation is the least understood of the three classes of particulate, although significant progress has been made in measuring ISP concentrations in operating recovery boilers and measuring the quantity of ISP formation during black liquor combustion of droplets and the char bed [Shaddix et al. 2004b]. Fundamental understanding gained by experimental work to-date may be sufficient to quantify the sources of ISP and to propose mechanisms for ISP formation and destruction in a recovery boiler, but it is not sufficient to develop sub-models to describe these processes.

Available sub-models for particle deposition are applicable over the entire range of particle size (0.1 to 1000 microns). However, there are still large uncertainties in the ability to predict fume deposition rates based on sub-models for thermophoretic deposition alone. The thermophoretic driving force decreases to near zero as deposits accumulate on surfaces and deposit surface temperature approaches the gas temperature. Both commercial operation and detailed laboratory experiments suggest that fume deposit mass increases proportionately with time and that other mechanisms such as “surface renewal” may be involved. A correlation for fume deposition rate was developed for a single tube in crossflow over the range of experimental conditions [Baxter et al. 2004a]; however, the correlation has not been tested for convection pass tubes of a recovery boiler. Submodels for deposit sintering and thermal properties depend on the ability to describe fume deposition accurately.

Comprehensive Model

Sub-models for particle deposition and deposit properties were implemented in B&W’s comprehensive model for recovery boilers, COMO-PR [Wessel and Baxter 2003; Wessel et al. 2004]. The model solves steady, three-dimensional turbulent flow, heat transfer, and black liquor combustion in a recovery boiler. The model predicts:

- combustion of black liquor spray and char deposits on the furnace walls and char bed;
- release of sodium, sulfur, potassium and chlorine;
- quantity and composition of inorganic material (including carryover, ISP, and submicron fume) leaving the furnace;
- distribution of gas velocity, temperature and chemical composition in the furnace and convection pass;

- distribution of convection and radiation heat transfer to the furnace walls and convection surfaces (tube banks);

Table 1. Status of Physics Sub-Model Development

Sub-Model	Maturity† (0 to 4)	Implemented in CFD Code	References
Particle Formation			Wessel & Verrill, 1998
Carryover*	3	✓	
Fume	2	✓	
ISP	1	✓	
Transformations/Reactions*	1		
Particle Deposition			Baxter & Sinquefield, 2004; Baxter, 2000; Wessel & Baxter, 2003
Inertial Impaction	3	✓	
Eddy Impaction*	2	✓	
Thermophoresis	3	✓	
Surface Renewal*	1		
Particle Capture/Sticking*	2	✓	
Deposit Sintering			Frederick, et al., Fuel, in press; Frederick, et al., Energy & Fuels, in press; Lunden, 1995.
Vaporization-Condensation*	2		
Solid-State Diffusion	1		
Deposit Structure*	1		
Deposit Strength	1		
Deposit Properties			Baxter, et al., 2001
Emissivity*	2	✓	
Thermal Conductivity*	2		
Tube Banks			Incropera & DeWitt, 1990
Radiation Heat Transfer	3	✓	
Convection Heat Transfer	3	✓	
Draft Loss	3	✓	

* Algorithms or correlations were developed or enhanced during this investigation

† Maturity of sub-model development based on experimental data, theory, algorithms, and model validation (0 – lowest, 4 - highest)

These modeling capabilities were previously described and demonstrated [Wessel et al. 1997; Verrill and Wessel 1998; Wessel and Verrill 1998; Wessel et al. 2000]. Sub-models for particle formation and deposition that were implemented in the comprehensive model were identified in Table 1 and described in the literature [Wessel and Baxter 2003]. However, as noted in the previous section, rational sub-models for ISP formation and destruction have not been developed, and sub-models are also needed to describe experimentally observed fume deposition rates and deposit properties. Without these sub-models, the ability to predict deposit formation and properties is limited.

Comprehensive modeling of black liquor combustion and ash deposition was performed for three recovery boilers listed in Table 2, for which boiler operating data, black liquor chemical analysis, and EP ash analysis were available. For two of these boilers, particle sampling and furnace probing data were also available for model validation.

Table 2. Comprehensive Recovery Boiler Modeling and Field Data

Unit Description	Firing Rate (lb/day virgin)	Particle Sampling	Furnace & Conv. Pass Probing Data
B&W recovery boiler in southern United States	3.25×10^6	none	none
Weyerhaeuser, Longview WA recovery boiler	4.83×10^6	Extractive samples, impaction probe samples, *in situ* laser-diagnostics	Gas velocity, temperature, and composition
International Paper, Courtland AL recovery boiler	3.88×10^6	Extractive samples, impaction probe samples, *in situ* laser-diagnostics	Gas velocity, and temperature (two ports only)

Modeling results for the southern U.S. boiler were described elsewhere [Wessel and Baxter 2003]. Modeling results for Longview and Courtland recovery boilers are presented in the ISP final project report [Shaddix et al. 2004b]. The reader is referred to these references for a discussion of the results and comparisons with field data. Modeling results were generally in good agreement with data, after anomalies in boiler operating data were rectified. However, ISP concentrations were substantially over predicted using the previously developed algorithm for ISP formation. More work is needed to characterize particle formation and deposition, and develop sub-models to describe these processes, before modeling can be used to postulate strategies to control fouling and plugging.

Conclusions

Field measurements were carried out to determine the characteristics and deposition of fume and ISP in recovery boilers and to quantify the size distribution of entrained particles and droplets in front of the superheaters and in the middle of the boiler bank. Fume particle size distributions peaked at about 0.7 μm physical diameter, and fume particle concentration ranged from 8.9 to 11.7 g/Nm^3 (normal conditions being 1 atm and 20ºC). Neither the size nor the concentration of fume particles changes significantly as the particles pass through the convection pass. Intermediate-sized particles (ISP) could not be reliably sampled, although estimates of their maximum concentration are as high as 50% of the total particle loading.

The thermal conductivity and emissivity of black liquor ash deposits were measured in laboratory experiments at conditions similar to actual fume deposits in a commercial recovery boiler. Theoretical models were developed to describe these properties in terms of deposit structure, porosity, and particle size. Experimental investigations of carryover deposition have significantly advanced our understanding and ability to predict large particle deposition in the convection pass of recovery boilers. Carryover is depleted in chloride and potassium compared to levels in the black liquor, which increases sticky temperature and reduces the tendency of these large particles to deposit on superheater tubes.

Studies of sintering and densification of fume deposits suggest there are no significant effects of gas environment on sintering rates, except at SO_2 concentrations of 1.0% which are unlikely to exist in the convection pass. An analysis of deposits formed in an operating recovery boiler revealed detailed information about the deposit structure and sintering behavior on tubes in the convection pass. A theoretical model for sintering was formulated, which incorporates different size-scales for sintering of poly-disperse particles and branched dendritic structures in low-density deposits. A simpler, yet more empirical form of sintering algorithms was also developed that is more tractable for engineering calculations.

Comprehensive modeling of particle formation and deposition was useful as a forensic tool for explaining experimental observations and evaluating particle formation and deposition sub-models. However, modeling has not yet been developed to the level of sophistication needed to predict the behavior of convection pass deposits accurately. Although ash deposition predictions are insightful, they cannot be analyzed or interpreted with confidence until rational sub-models for ISP formation and destruction are developed. Sub-models are also needed to describe experimentally observed fume deposition rates, deposit properties, and potential interaction with ISP particles. Field data gathered on this project will be valuable for testing new algorithms, for validating modeling results, and for improving our understanding of particle formation and deposition processes.

Technology Transfer

The results of this project were disseminated to the pulp and paper industry in a number of ways during the course of this project.

- Interim results of this project were presented on four occasions at the University of Toronto, Pulp & Paper Center, Annual Research Review Meeting, Toronto, Ontario, Canada in November 1998, 1999, 2000, and 2001. Participants of these meetings included consortium members from several pulp and paper companies, equipment suppliers, and industry consultants. These meetings provided valuable feedback from industry which helped guide the direction of this project.

- The status and results of this project were also presented annually to the American Forest and Paper Association (AF&PA) Agenda 2020 Energy Performance Committee.

- A conference was organized by Larry Baxter (Sandia), Rick Wessel (B&W), and Esa Vakkilainen (Ahlstrom Machinery), and sponsored by the United Engineering Foundation, that focused on issues directly related to this project. The conference was titled "Behavior of Inorganic Material in Recovery Boilers" and was held in Bar Harbor Maine, June 4-9, 2000. There were 54 participants from research and industry, spanning seven countries and three continents (North America, Europe and Asia). A total of 42 technical papers were presented (five from this project). The outcome of the conference was a loose-bound folder of technical papers/presentations that was distributed to participants, and a list of key issues and needs that were identified during discussions by participants and summarized in a consensus white paper.

- Results of this project are documented in several technical publications, authored by the principal investigators or their colleagues. These papers are referenced throughout this report and included in the List of References.

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